

## \* NOTICES \*

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## DETAILED DESCRIPTION

## [Detailed Description of the Invention]

[0001]

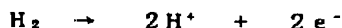
[Industrial Application] This invention relates to the electrode for polymer electrolyte fuel cells, and the polymer electrolyte fuel cell using it especially about the fuel cell which uses reducing agents, such as pure hydrogen or a methanol, and reforming hydrogen from a fossil fuel, and oxidizers, such as air and oxygen, as a fuel.

[0002]

[Description of the Prior Art] The polymer electrolyte fuel cell (PEFC) uses for the electrolyte the ion exchange membrane which is a solid-state polyelectrolyte, and when hydrogen is used as a fuel, the reaction of (\*\* 1) occurs in a negative electrode.

[0003]

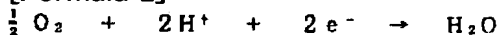
[Formula 1]



[0004] Moreover, when oxygen is used as an oxidizer, in a positive electrode, the reaction of (\*\* 2) occurs and water is generated.

[0005]

[Formula 2]



[0006] PEFC is a fuel cell of high performance with which two or more 1 A/cm high power is obtained by ordinary temperature and ordinary pressure. The electrode design in consideration of formation of the gas channel which supplies the increment in the touch area of the Pt particle and the solid-state polyelectrolyte which are an electrode catalyst, i.e., reaction area, and reactant gas for realizing this high power becomes important.

[0007] In the case of the electrode for phosphoric acid fuel cells (PAFC) which uses for an electrode the catalyst which supported noble metals similar to PEFC to carbon, by J. Electroanal. Chem. and 195 (1985) 81, the electrolytic solution is held at detailed pore with a diameter of 0.1 micrometers or less, and it is supposed that the pore with a larger diameter than 0.1 micrometers is become the supply way of reactant gas. In JP, 6-267545, A, as for volume of 10-100 micrometers, the pore diameter of 0.1 micrometers or less of a catalyst bed and 11% or more of volume of 0.1-1.0 micrometers are confirmed 42% or less as a positive electrode of a phosphoric acid fuel cell, respectively.

[0008]

[Problem(s) to be Solved by the Invention] However, in the case of PEFC, since the high macromolecule of \*\* is used for the electrolyte with the molecular level, it is thought that the reaction place formed differs from the fuel cell using the electrolytic solution like the phosphoric acid which is low-molecular [above-mentioned / conventional]. For this reason, said JP, 6-267545, A is the thing of the electrode for phosphoric acid fuel cells, and does not serve as a design manual of the electrode for PEFC. Moreover, examination about the pore structure of the

electrode which fitted PEFC until now was not performed. For this reason, in order to realize more highly efficient PEFC, it was required for the supply ability of reactant gas to acquire the optimal pore structure of the high electrode for PEFC greatly [ the touch area of Pt catalyst and a solid-state polyelectrolyte ].

[0009] This invention aims at offering PEFC using the highly efficient electrode for PEFC and highly efficient it by solving the above-mentioned technical problem, considering pore distribution of the catalyst bed of the electrode for PEFC, and acquiring the optimal pore structure of the electrode for PEFC.

[0010]

[Means for Solving the Problem] The electrode for polymer electrolyte fuel cells of this invention is an electrode which formed in one side of a gaseous diffusion layer the catalyst bed which consists of a solid-state polyelectrolyte and carbon powder which supported the precious metal catalyst at least, and the specific volume of pore with a diameter [ in said catalyst bed ] of 0.04–1.0 micrometers carries out it to more than 0.06m<sup>3</sup>/g preferably more than 0.04cm<sup>3</sup>/g.

[0011] Furthermore, it is the polymer electrolyte fuel cell which used said electrode at least for one side of the electrode arranged on both sides of the solid-state polyelectrolyte film.

[0012]

[Function] In the electrode for PEFC, it can be said that the solid-state polyelectrolyte is distributed over the pore section with a diameter of 0.04–1.0 micrometers. That is, it is thought that this pore section functions as a reaction place. Moreover, it is thought that this pore section functions as a supply way (gas channel) of the reactant gas of hydrogen and oxygen as well as the case of a phosphoric acid fuel cell. Therefore, in PEFC, it can be said that a reaction place exists in the pore section with a diameter of 0.04–1.0 micrometers which functions also as a gas channel.

[0013] For this reason, the more highly efficient electrode for PEFC and more highly efficient PEFC can be offered by searching for the pore distribution large the touch area of Pt catalyst and a solid-state solid-state polyelectrolyte, i.e., reaction area, and optimal for PEFC to which the supply ability of reactant gas becomes high.

[0014]

[Example] Hereafter, the example of this invention is explained, referring to a drawing.

[0015] (Example 1) As an alcoholic solution of a solid-state solid-state polyelectrolyte, mixed churning of the 5%Nafion solution by the Aldrich chemical company was carried out with n-butyl acetate so that the amount of solid-state solid-state polyelectrolytes might serve as 0.1 – 1.4 mg/cm<sup>2</sup>, and the colloid dispersion liquid of four kinds of polyelectrolytes were generated. The carbon powder which made these colloid dispersion liquid support Pt catalyst 20 to 30% of the weight was added, and colloid was made to stick to the front face of the carbon powder which made Pt catalyst support. These dispersion liquid were made into the shape of a paste using the ultrasonic distribution machine. This paste was applied on the carbon paper beforehand covered with 30 – 60% of the weight of the fluororesin, and the electrode of this invention was produced.

[0016] In order to clarify pore structure of the electrode to which the amount of these solid-states polyelectrolytes was changed, pore distribution was measured with the method of mercury penetration.

[0017] Change of the pore distribution at the time of using Pt support carbon powder for drawing 1, and changing the amount of solid-state polyelectrolytes of an electrode is shown. Drawing shows that the pore specific volume (differential value) of the diameter the part of 0.04–1.0 micrometers which has a peak by change of the amount of solid-state polyelectrolytes is changing. In addition, although the pore specific volume of 1.0 micrometers or more is also changing, this part is the pore section resulting from carbon paper.

[0018] The relation of the pore specific volume which is in the amount of solid-state polyelectrolytes and the diameter of 0.04–1.0 micrometers at drawing 2 is shown. From drawing, since the pore specific volume in the diameter of 0.04–1.0 micrometers decreases by the increment in a solid-state polyelectrolyte, it can be said that the solid-state polyelectrolyte was distributed over this pore section.

[0019] That is, it is thought that this pore section functions as a reaction place. Moreover, it is thought that the pore in this diameter of 0.04–1.0 micrometers functions as a supply way (gas channel) of the reactant gas of hydrogen and oxygen like the case of a phosphoric acid fuel cell. Therefore, in PEFC, it can be said that a reaction place exists in the pore section with a diameter of 0.04–1.0 micrometers which functions also as a gas channel.

[0020] (Example 2) The electrode was produced by the same approach as an example 1. At this time, nine kinds of carbon powder with which specific surface area differs from the diameter of a primary particle as carbon powder which is the support of Pt catalyst was used. Moreover, the amount of solid-state polyelectrolytes was made into 1.0 mg/cm<sup>2</sup>.

[0021] The hotpress of this electrode was carried out to both sides of the Du Pont solid-state polyelectrolyte film Nafion115 by the temperature of 120–200 degrees C, and the pressure of 50–100kg/cm<sup>2</sup>, and the cell of this invention was produced.

[0022] Pore distribution of an electrode was measured with the method of mercury penetration. Moreover, the spark test of a cell was performed at ordinary pressure and the cell temperature of 50 degrees C, using hydrogen–oxygen as a fuel.

[0023] The relation between the pore specific volume which is in the diameter of 0.04–1.0 micrometers of the electrode of this example at drawing 3, and the current density in 850mV of a cell is shown. Consequently, the current density which can be taken out with the increment in pore specific volume increased. Since the current density in 850mV which is the rule region of activation polarization is increasing, it can be said that reaction area was increased with pore specific volume.

[0024] In addition, the carbon of Point A has deviated from this straight line. The solid-state polyelectrolyte addition of an electrode catalyst bed is because, as for this, reaction area decreased by the continuity of a solid-state polyelectrolyte falling with 1.0 mg/cm<sup>2</sup> since it is fixed, and pore specific volume is large. If the amount of solid-state polyelectrolytes is optimized as 1.5 mg/cm<sup>2</sup>, reaction area will be made to increase in connection with pore specific volume becoming large, current density will also increase, and it will get on a straight line. (Point A').

[0025] The relation between pore specific volume with a diameter [ of the electrode of this example ] of 0.04–1.0 micrometers and the electrical–potential–difference value in 400 mA/cm<sup>2</sup> of a cell is shown in drawing 4. Pore specific volume showed the high electrical potential difference above 0.04cm<sup>3</sup>/g, and the electrical potential difference became almost fixed above 0.06cm<sup>3</sup>/g.

[0026] The relation between pore specific volume with a diameter [ of the electrode of this example ] of 0.04–1.0 micrometers and the electrical–potential–difference value in 800 mA/cm<sup>2</sup> of a cell is shown in drawing 5. Pore specific volume showed high cell voltage above 0.06cm<sup>3</sup>/g. On the other hand, in the range of 0.04–0.06cm<sup>3</sup>/g, as for the cell voltage to pore specific volume, variation was seen by the cell.

[0027] In 400 and 800 mA/cm<sup>2</sup>, since concentration polarization becomes dominant, the supply ability of not only reaction area but reactant gas becomes important. Since the pore in the diameter of 0.04–1.0 micrometers functions also as a gas channel, since gas supply ability is low, pore specific volume is considered that cell voltage became low below by 0.04cm<sup>3</sup>/g.

[0028] Moreover, in a high current density region, it becomes important [ the discharge ability of generation water ] at gas supply ability and coincidence. For this reason, the variation in the electrical potential difference by the cell in the range of 0.04–0.06cm<sup>3</sup>/g in 800 mA/cm<sup>2</sup> is considered because the discharge ability of the generation water by the hydrophilic property/hydrophobicity of each carbon powder differs. Therefore, in order not to make it dependent on the property of carbon powder, the pore specific volume more than 0.06cm<sup>3</sup>/g is needed in a high current density region.

[0029] In addition, although 5%Nafion solution by U.S. Aldrich Chemical was used as a solid-state polyelectrolyte in this example as an example of representation of the giant molecule which consists of a copolymer of tetrafluoroethylene and perfluoro vinyl ether It is not what will be limited to the above–mentioned example if it is a solid-state polyelectrolyte with a proton exchange group. the molecular structure — things — the same effectiveness was acquired even if it used the giant molecule with which a giant molecule, for example, perfluoro vinyl ether, differs

from side-chain molecule length, and the giant molecule which consists of a copolymer of styrene and vinylbenzene.

[0030] Moreover, the electrode producing method of this example shows an example, and is not limited to this.

[0031] Furthermore, although the hydrogen-oxygen fuel cell which used the solid-state polyelectrolyte film for the electrolyte as an example of a fuel cell was taken up in this example, it is also possible to apply a methanol, natural gas, naphtha, etc. to the fuel cell using the reforming hydrogen used as a fuel or the fuel cell using air as an oxidizer.

[0032]

[Effect of the Invention] According to this invention, the supply ability of reactant gas can realize PEFC using the more highly efficient high electrode for PEFC and more highly efficient high it greatly [ a touch area with the solid-state polyelectrolyte of Pt catalyst ] by clarifying pore structure suitable for PEFC as mentioned above.

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[Translation done.]

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**DESCRIPTION OF DRAWINGS**

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[Brief Description of the Drawings]

[Drawing 1] Drawing showing the amount of solid-state polyelectrolytes of the electrode of the example of this invention, and the relation of pore distribution

[Drawing 2] Drawing showing the amount of solid-state polyelectrolytes of the electrode of the example of this invention, and the relation of pore specific volume

[Drawing 3] Drawing showing the relation between the pore specific volume of the electrode of the example of this invention, and a cell property (the 1)

[Drawing 4] Drawing showing the relation between the pore specific volume of the electrode of the example of this invention, and a cell property (the 2)

[Drawing 5] Drawing showing the relation between the pore specific volume of the electrode of the example of this invention, and a cell property (the 3)

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[Translation done.]